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(54) Title: PLANT OIL AND CHEMICAL COMPOUND HAVING ACARICIDAL ACTIVITY

(57) Abstract: The present invention relates to an acaricidal composition. In particular, the present invention relates to acaricidal composition, comprising plant oils extracted from member selected from at least one group consisting of *Pimpinella anisum*, *Laurus nobilis*, *Melaleuca leucadendron*, *Elettaria cardamomum*, *Pseudotsuga menziesii*, *Foeniculum vulgare*, *Ferula galbaniflua*, *Pelargonium odoratissimum*, *Pelagonium radens*, *Pelagonium capitatum*, *Helichrysum augustifolium*, *Andropogon muricatus*, *Lavendula officinalis*, *Origanum majorana*, *Milissa officinalis*, *Citrus aurantium*, *Melaleuca viridiflora*, *Ravensara aromatica*, *Sassafras albidum*, *Tagetes erecta*, *Tagetes patula*, *Verbena officinalis*, *Thujopsis dolabrata* and monoterpene compounds selected at least one group consisting of carvacrol, (+)-fenchol, geraniol, linanol, (-)-cis-myrtanol, trans-myrtanol, (-)-myrtenal, (-)-myrtenol, thujone, cis-verbenol, (-)-verbenone, menthone, eugenol, menthol, and indoline. The present invention shows potent acaricidal activities against mites as well as superior effect to conventional synthetic acaricide, therefore, said plant oils and compounds are expected to be useful as acaricidal composition.

PLANT OIL AND CHEMICAL COMPOUND HAVING ACARICIDAL ACTIVITY

Technical Field

5 The present invention relates to an acaricidal composition containing an extract of plant or a monoterpene compound isolated therefrom.

Background Art

10 Various allergens are known which are transported through the air to trigger a human reaction. For example, it has been known for a long time that house dust can trigger allergic reactions in humans. It was reported, as early as 1928 that the mites in the dust were the primary source of the allergic response, but it was only in the 1960's that researchers appreciated its significance.

15 It is believed that the feces of the dust mite trigger the immune response of the body, thereby giving rise to well known allergic symptoms, such as asthma and rhinitis (Maunsell, K. et al; *Lancet* 1, p1267-1270, 1968). The mite belonging to Pylogryphidae family inhabits in dust and *Dermatophagoides pteronyssinus* (known as Der-p), *Dermatophagoides farinae* (known as Der-f), *Dermatophagoides microceras*, *Euroglyphus maynei* et al. causes the human
20 having physically atopic constitution to an asthma or eczema and the like disease (See Voorhorst, R. et al; *Asthma*, 10(6), p329-334, 1964; International Workshop Report. *The Journal of Allergy and Clinical Immunology*, 83, p416-427, 1989; and *bid*, 89, p1046-1060, 1992).

 The most of mites found in Korea are dust mites i.e., Def-p, Der-f species found in carpet,
25 clothes, furniture, mats and bedclothes etc. (De Boer, R.; *The acari: reproduction, development, and life history strategies*, NY. Chapman and Hall, p517-518, 1991). The dust mites hate sunlight and require an appropriate temperature and humidity condition for proliferation of

offspring and human corneous tissue detached from dried skin for a source of food. Therefore, to kill the mites, various methods comprising cleaning all the dust in room, sterilizing household goods, clothes, carpet and the like at the temperature of more than 70°C and controlling a temperature and humidity have been studied till now. But the above methods have lots of barrier and difficulty in obtaining complete exterminating spectrum for the mites.

The compounds having acaricidal activity are known as lindane, pirimiphos-methyl, benzyl benzoate, dibutyl phthalate and diethyl *m*-toluamides (Hellet-Haupt A. and Busvine, J. R.; *J. Med. Entomol.*, 2(5), p551-558, 1974). But the use of lindane compound as a household acaricide is limited because of its high toxicity although the compound has most effective acaricidal activity and consistency.

Chemical control using by insecticide has been conducted to get rid of mites, however, the control causes to drug tolerance for some species of the mites, for example, some mites belonging to Pyroglyphid family showing drug tolerance for DDT and lindane, are found (Bronsjik, J.E.M.H. et al; *Res. Popul. Ecol.*, 8, p67-79, 1971)

Therefore, many attempts are tried to substitute the conventional acaricide in the world. Especially, natural acaricide using by plant extract and aromatic compound has been studied extensively (Miyazaki, Y. et al; *Jpn. J. Biometeor.* 26 p105-108, 1989; Watanabe, F. et al; *Shoyakugaku Zasshi* 43 p163-168, 1989; Yatagai, M. and Morita, S.: *J. Japan Wood Res. Soc.* 37, p345-351, 1991).

The present inventors have endeavored to develop a plant extract and aromatic compounds showing high acaricidal activity, and have found that various plant extract and compounds show potent acaricidal activities against some species of mites.

Disclosure of Invention

An object of the present invention is therefore to provide an extract of plant having potent acaricidal activity.

Another object of the present invention is to provide a monoterpene compound

having acaricidal activity.

In an embodiment of the present invention, there is provided an acaricidal composition comprising plant oils extracted from member selected from at least one group consisting of *Pimpinella anisum*, *Laurus nobilis*, *Melaleuca leucadendron*, *Elettaria cardamomum*,
5 *Pseudotsuga menziesii*, *Foeniculum vulgare*, *Ferula galbaniflua*, *Pelargonium odoratissimum*, *Pelargonium radens*, *Pelargonium capitatum*, *Helichrysum angustifolium*, *Andropogon muricatus*, *Lavendula officinalis*, *Origanum majorana*, *Milissa officinalis*, *Melaleuca viridiflora*, *Ravensara aromatica*, *Sassafras albidum*, *Tagetes erecta*, *Tagetes patula*, *Verbena officinalis* and *Thujopsis dolabrata*.

10 In another embodiment of the present invention, there is also provided a process for preparing plant oil from above mentioned plants characterized in subjecting above plants to successive-vapor evaporation method.

For example, above plant is sliced and dried in the shade. Then, together with one to twenty times, preferably, three to ten times volumes of water or $C_1 \sim C_5$ lower alcohol, preferably, methanol, ethanol or isopropanol and about 0.5 to 2 times, preferably, 0.6 to 1 time
15 volumes of nonpolar solvent preferably, hexane, ethyl-acetate, acetone, more preferably, hexane solvent than the weight of dried plant (V/W) is added to the dried plant and the mixture is subjected to successive-vapor evaporation extraction, percolation extraction, ultra-sonication extraction, reflux-cooling extraction or soxhlet extraction, preferably, successive-vapor
20 evaporation extraction for a period ranging from 1 hour to 48 hours, preferably from 5 hours to 24 hours, at a temperature ranging from 0 to 150°C, preferably 10 to 100 °C. The extract is filtered, dried with drying agent, preferably, anhydrous magnesium sulfate, anhydrous calcium sulfate, anhydrous potassium sulfate, anhydrous silica gel, more preferably, anhydrous magnesium sulfate, concentrated under a reduced pressure at temperature ranging from 0 to
25 40°C, preferably, from 4 to 20 °C and the concentrates is sealed and kept in refrigerator.

In another embodiment, the compositions of the present invention include at least one terpene. Non-limiting examples of terpenes, which can be found in the compositions of the

present invention, include cavacrol, (+)-fenchol, geraniol, linanol, (-)-*cis*-myrtanol, *trans*-myrtanol, (-)-myrtenal. (-)-myrtenol, thujone, *cis*-verbenol, (-)-verbenone, menthone, eugenol, menthol and indoline, which can be isolated from said plant extract or purchased at any market place.

5 The monoterpenes and the sesquiterpenes are the main components of essential oils. Monoterpenes, derived from 2 isoprene units, are 10-carbon compounds (C, 10) and the sesquiterpenes, derived from 3 isoprene units are 15-carbon compounds (C, 15). Both mono and sesquiterpenes belong to the steam distillate fraction i.e. essential oils, because of their low boiling point. These compounds are known to be present in fragrant, volatile, essential oils of
10 many plants including mints, pine, cedar, citrus, eucalyptus and spices.

 The acaricidal composition of present invention comprises said plant oils or monoterpene compounds in an amount of 0.01 to 40 % (w/w), preferably, 0.1 to 20 % (w/w) with a sole or mixture type in the amount of total composition. However, the amount of said oil or compound could be modified according to the preparation of insecticide, spreading method,
15 the area of spreading and the species of insecticide.

 The inventive composition is used wherein mite inhabits, the preferable application methods are spreading treatment, decoction treatment, fumigation treatment and the like, the dosage of application is the amount of conventional dosage of insecticide. The preferable formulations of said composition are liquid, emulsion, smoke, fumigation, spray, granule, solid
20 and the like types.

 The inventive composition may additionally comprise conventional carrier in accordance with a using method. It is preferable that said carrier is used as a appropriate substance according to the usage and application method.

 The inventive composition comprises an insecticidal composition having acaricidal
25 activity specifically against mite.

 Specifically, the inventive composition is effective in Der-p or Der-f.

 Additionally, it is preferable that said composition is applied as a form of volatile

toxicant or fumigate.

Best Mode for Carrying Out the Invention

5 The following Example and Test Example are intended to further illustrate the present invention without limiting its scope.

Example 1: Isolation of plant oils from plants having acaricidal activity

10 Acaricidal plants i.e., of *Pimpinella anisum*, *Laurus nobilis*, *Melaleuca leucadendron*, *Elettaria cardamomum*, *Pseudotsuga menziesii*, *Foeniculum vulgare*, *Ferula galbaniflua*, *Pelargonium odoratissimum*, *Pelargonium radens*, *Pelargonium capitatum*, *Helichrysum augustifolium*, *Andropogon muricatus*, *Lavendula officinalis*, *Origanum majorana*, *Milissa officinalis*, *Melaleuca viridiflora*, *Ravensara aromatica*, *Sassafras albidum*, *Tagetes erecta*,
15 *Tagetes patula*, *Verbena officinalis* and *Thujopsis dolabrata* were cropped in USA, Australia and Korea. The 23 species of plants were dried in the shade, minced with a slice and weighed at 300g respectively. 2000 ml of distilled water and 200 ml of hexane were poured to the sliced plants and the mixture was subjected to successive-vapor evaporation extraction with SDE apparatus (Nickerson & Likens) at 100 °C, for 6 hours. The extract was dried with anhydride
20 magnesium sulfate, concentrated under a reduced pressure, sealed and kept in refrigerator.

Example 2: Preparation of monoterpene compounds having acaricidal activity

 Acaricidal compounds i.e., carvacrol (catalog No. 22051), (+)-fenchol (cataog No.
25 46198), geraniol (catalog No. 48798 or 48799), (-)-*cis*-myrtanol (catalog No. 70154), *trans*-myrtanol (catalog No. 70117 or 70155), (-)-myrtenal (catalog No. 70119 or 70125), (-)-myrtenol (catalog No. 70158), thujone (catalog No. 89230), *cis*-verbenol (catalog No., 94879),

(-)-verbenone (catalog No. 94882), menthone (catalog No. 63680), eugenol (catalog No. 35995 or 46100), menthol (catalog No., 63670 or 63671) and indoline (catalog No. 57240) were purchased from Fluka Co. located in Switzerland and linanol (Fluca: catalog No. 62140, Wako: catalog No. 126-00993) was purchased from Wako Co. located in Japan.

5

Test Example 1: Acaricidal activities of the plant oils in example 1.

The acaricidal activities of the plants oil in example 1 against various mites were determined by the procedure described in a literature (Watanabe, F. et al; *Shoyakugaku Zasshi* 10 43, p163-168, 1989; Yatagai, M. and Morita, S.; *J. Japan Wood Res, Soc.* 37, p345-351, 1991).

The preparation of Mites

Two species of mites, i.e., European house dust mite (*Dermatophagoides pteronyssinus*) and American house dust mite (*Dermatophagoides farinae*) were bred in 15 breeding room maintaining at relative humidity ($75 \pm 5\%$) and temperature ($25 \pm 1^\circ\text{C}$) and the mixture feed of fry feed and complex vitamin (Ebiose[®], Samil Pharm) was fed in a vessel (12.5 x 10.5 x 5 cm) and an appropriate saline solution was poured at the bottom of another vessel (17.5 x 17.5 x 17.5 cm). The mites were bred without exposing any insecticide.

20 The determination of acaricidal activity

Most important point among screening steps to identify biologically active compounds is setting an initial concentration of sample. In present invention; since the characteristic of mite is microscopic, the concentration was determined 1 mg of sample per 2 ml of microtube through repeated preliminary experiments. Various concentrations of plant oils (1.0, 0.5, 0.25, 25 0.125 and 0.062 mg each) were dissolved in 40 μl of methanol, each solution was poured in 2 ml of colorless microtube and the tube was stirred in order to the solution containing each sample can be spread and adhered to wall and tip of the tube thoroughly. And then, the

methanol was removed in freeze dryer for 1 hour. 25 numbers of mites were added to each tube and tested at the temperature of $25 \pm 1^\circ\text{C}$. The mites were selected from observing the activities of mites with microscope. The result was tested after 24 hours by microscopic examination (x 20) and determined by the criteria that motionless mites were regarded as dead one when stimulating appendage and body by minute brush. All test were repeated at 5 times and the comparison of average value between acaricidal rates was calculated by formula of Scheffe' test (SAS Institute, 1996). Control group was treated with same procedure without plant oil.

The acaricidal rate of plant oils in example 1 was shown in Table 1 and 2, wherein identical letter i.e., "a" and "b" means that the average showed no significant difference statistically ($P = 0.05$, Scheffe' test) and the rate of insecticide was shown by transforming to arcsine square root.

[Table 1] Acaricidal activities of the plant oils against *Dermatophagoides pteronyssinus*

The Species of Plants	Acaricidal rate (Average \pm Standard Deviation, %)				
	The Concentration of treated sample (mg/tube)				
	1.00	0.50	0.250	0.125	0.062
<i>Pimpinella anisum</i>	100a	100a	100a	75.3 \pm 1.7b	14.3 \pm 1.9c
<i>Laurus nobilis</i>	100a	100a	100a	100a	56.5 \pm 1.3b
<i>Melaleuca leucadendron</i>	100a	100a	100a	31.6 \pm 2.1b	0c
<i>Elettaria cardamomum</i>	100a	100a	100a	100a	32.3 \pm 1.7b
<i>Pseudotsuga menziesii</i>	100a	100a	100a	15.2 \pm 1.5b	0c
<i>Foeniculum vulgare</i>	100a	100a	70.3 \pm 1.5b	10.2 \pm 1.1c	0d
<i>Ferula galbaniflua</i>	100a	100a	100a	85.9 \pm 1.2b	25.2 \pm 2.4c
<i>Pelargoniummordoratissimum</i>	100a	100a	100a	100a	53.1 \pm 1.8b
<i>Pelagonium radens</i>	100a	100a	100a	100a	26.7 \pm 2.1b
<i>Pelagonium capitatum</i>	100a	100a	100a	100a	36.4 \pm 2.3b

<i>Helichrysum augustifolium</i>	100a	100a	100a	100a	0b
<i>Andropogon muricatus</i>	100a	100a	100a	100a	53.4±1.8b
<i>Lavendula officinalis</i>	100a	100a	100a	92.4±2.1b	0c
<i>Origanum majorana</i>	100a	100a	100a	26.3±2.6b	0c
<i>Milissa officinalis</i>	100a	100a	91.8±1.9b	14.3±1.4c	0d
<i>Citrus aurantium</i>	100a	100a	100a	100a	26.2±1.4b
<i>Melaleuca viridiflora</i>	100a	100a	100a	91.7±2.6b	32.3±1.8c
<i>Ravensara aromatica</i>	100a	100a	34.5±2.0b	0c	0c
<i>Sassafras albidum</i>	100a	100a	95.2±1.0b	13.5±1.1c	0d
<i>Tagetes erecta</i>	100a	100a	100a	100a	100a
<i>Tagetes patula</i>	100a	100a	100a	100a	100a
<i>Verbena officinalis</i>	100a	100a	100a	100a	58.9±2.1b
<i>Thujopsis dolabrata</i>	100a	100a	100a	100a	69.2±1.2b

[Table 2] Acaricidal activities of the plant oils against *Dermatophagoides farinae*

The Species of Plants	Acaricidal rate (Average±Standard Deviation, %)				
	The Concentration of treated sample (mg/tube)				
	1.00	0.50	0.250	0.125	0.062
<i>Pimpinella anisum</i>	100a	100a	100a	64.4±1.3b	9.5±1.4c
<i>Laurus nobilis</i>	100a	100a	100a	100a	50.3±1.1b
<i>Melaleuca leucadendron</i>	100a	100a	100a	25.2±1.5b	0c
<i>Elettaria cardamomum</i>	100a	100a	100a	100a	20.5±1.1b
<i>Pseudotsuga menziesii</i>	100a	100a	100a	11.2±1.2b	0c
<i>Foeniculum vulgare</i>	100a	100a	65.4±1.2b	0c	0c
<i>Ferula galbaniflua</i>	100a	100a	100a	81.2±1.1b	22.3±1.8c

<i>Pelargoniumordoratissimum</i>	100a	100a	100a	100a	50.6±1.5b
<i>Pelagonium radens</i>	100a	100a	100a	100a	20.4±1.8b
<i>Pelagonium capitatum</i>	100a	100a	100a	100a	29.8±1.7b
<i>Helichrysum augustifolium</i>	100a	100a	100a	100a	0b
<i>Andropogon muricatus</i>	100a	100a	100a	100a	51.4±1.5b
<i>Lavendula officinalis</i>	100a	100a	100a	90.8±1.5b	0c
<i>Origanum majorana</i>	100a	100a	100a	22.5±1.7b	0c
<i>Milissa officinalis</i>	100a	100a	90.5±1.5b	12.1±1.9c	0d
<i>Citrus aurantium</i>	100a	100a	100a	100a	23.5±1.5b
<i>Melaleuca viridiflora</i>	100a	100a	100a	89.8±2.1b	28.9±1.5c
<i>Ravensara aromatica</i>	100a	100a	31.6±1.5b	0c	0c
<i>Sassafras albudum</i>	100a	100a	93.8±1.8b	12.1±1.7c	0d
<i>Tagetes erecta</i>	100a	100a	100a	100a	100a
<i>Tagetes patula</i>	100a	100a	100a	100a	100a
<i>Verbena officinalis</i>	100a	100a	100a	100a	53.3±1.5b
<i>Thujopsis dolabrata</i>	100a	100a	100a	100a	68.4±1.9b

As can be seen in Table 1 showing the result of acaricidal activities tested in *Dermatophagoides pteronyssinus*, all the 23 plant oils in Example 1 showed 100% of acaricidal rate at the concentration of each 1 mg and 0.5 mg, in the concentration of 0.125 mg, the plant oils from *Pimpinella anisum*, *Laurus nobilis*, *Elettaria cardamomum*, *Ferula galbaniflua*, *Pelargonium ordoratissimum*, *Pelagonium radens*, *Pelagonium capitatum*, *Helichrysum augustifolium*, *Andropogon muricatus*, *Lavendula officinalis*, *Citrus aurantium*, *Melaleuca viridiflora*, *Sassafras albudum*, *Tagetes erecta*, *Tagetes patula*, *Verbena officinalis* and *Thujopsis dolabrata* showed more than 70% of acaricidal rate. However, in the concentration of 0.062 mg, the plant oils from *Laurus nobilis*, *Pelargonium ordoratissimum*, *Andropogon muricatus*, *Verbena officinalis* and *Thujopsis dolabrata* only showed more than 50% of

acaricidal rate.

As can be seen in Table 2 showing the result of acaricidal activities tested in *Dermatophagoides farinae*, all the 23 plant oils in Example 1 showed 100% of acaricidal rate all the concentration of 1 mg and 0.5 mg, in the concentration of 0.062 mg, the plant oils from

5 *Laurus nobilis*, *Pelargonium odoratissimum*, *Andropogon muricatus*, *Verbena officinalis* and *Thujopsis dolabrata* only showed more than 50% of acaricidal rate.

Test Example 2: Acaricidal activities of the monoterpene compounds in example 2.

10 The acaricidal activities of the monoterpene compounds in example 2 against various mites were determined as follows.

The determination of acaricidal activity

Identical determination methods with those of Test Example 1 were subjected to the

15 monoterpene compounds i.e., carvacrol, (+)-fenchol, geraniol, linanol, (-)-*cis*-myrtanol, *trans*-myrtanol, (-)-myrtenal, myrtenol, thujone, *cis*-verbenol, (-)-verbenone, menthone, eugenol, menthol and indoline in Example 2.

Table 3 showed acaricidal activities of monoterpene compounds in Example 1 against American house dust mite (*Dermatophagoides farinae*) and Table 4 showed European house

20 dust mite (*Dermatophagoides pteronyssinus*) respectively.

[Table 3] Acaricidal activities of monoterpene compounds against *Dermatophagoides pteronyssinus*

Compound	Acaricidal rate (Average±Standard Deviation, %)						
	The Concentration of treated sample (mg/tube)						
	0.50	0.33	0.25	0.10	0.050	0.025	0.010

						2.1b	1.8c
Indoline	100a	100a	100a	100a	25.2± 1.8b	0c	0c
Benzyl Benzoate	100a	100a	88.8± 3.6b	29.5±1.5c	0d	0d	0d
Dibutyl phtalate	100a	100a	95.2± 1.4a	54.4±3.3b	27.4± 1.9c	0d	0d

As can be seen in Table 3, all the 15 compounds from Example 2 showed 100% of acaricidal activities in *Dermatophagoides pteronyssinus* at the concentration of 0.5 mg, 0.33 mg and 0.25 mg. At the concentration of 0.010 mg, carvacrol, geraniol, linanol, thujone, eugenol and menthol showed acaricidal activities 73.8%, 76.7%, 36.2%, 35.3%, 23.4%, and 43.3%, respectively. These facts proved the monoterpene compounds have superior effect to control groups, i.e., benzyl benzoate and dibutyl phthalate.

[Table 4] Acaricidal activities of monoterpene compounds against *Dermatophagoides farinae*

10

[illegible]

						1.1b	1.5c
(-)- <i>cis</i> -myrtanol	100a	100a	100a	79.2± 1.7b	42.3± 2.0c	0d	0d
<i>trans</i> -myrtanol	100a	100a	100a	78.4± 2.1b	48.5± 1.8c	0d	0d
(-)-myrtenal	100a	100a	100a	100a	100a	46.7± 1.6b	0c
(-)-myrtenol	100a	100a	100a	76.5± 1.6b	40.6± 1.4c	0d	0d
Thujone	100a	100a	100a	100a	76.5± 1.3b	36.4± 2.9c	0d
<i>cis</i> -verbenol	100a	100a	100a	100a	84.2± 2.2b	26.5± 1.4c	0d
(-)-verbenone	100a	100a	100a	100a	100a	26.9± 1.8b	0c
Menthone	100a	100a	100a	65.3± 1.5b	28.7± 2.5c	0d	0d
Eugenol	100a	100a	100a	100a	100a	76.7± 2.5b	36.6± 1.3b
Menthol	100a	100a	100a	100a	100a	82.8± 1.9b	34.1± 1.6c
Indoline	100a	100a	100a	100a	20.7± 1.6b	0c	0c
Benzyl Benzoate	100a	100a	100a	32.0± 2.9b	0c	0c	0c
Dibutyl phthalate	100a	100a	92.0± 3.8b	62.2± 1.9c	29.9± 1.7d	0e	0e

As can be seen in Table 4, all the 15 compounds from Example 2 showed 100% of acaricidal rate in *Dermatophagoides farinae* at the concentration of 0.5 mg, 0.33 mg and 0.25 mg. At the concentration of 0.010 mg, carvacrol, geraniol, linanol, eugenol and menthol showed acaricidal activities 76.7%, 83.3%, 43.2%, 36.6%, 34.1%, respectively. However, the control groups, i.e., benzyl benzoate and dibutyl phtalate did not show any significant acaricidal activities at the concentrations of less than 0.050 mg.

Industrial Applicability

As mentioned above, the plant oils and monoterpene compounds of present invention showed potent acaricidal activities against mite as well as superior effect to conventional synthetic acaricide, therefore, said plant oils and compounds are expected to be useful as acaricidal composition.

While the invention has been described with respect to the above specific embodiments, it should be recognized that various modifications and changes may be made to the invention by those skilled in the art which also fall within the scope of the invention as defined in the appended claims.

Claims

1. An acaricidal composition comprising plant oils extracted from member selected from at least one group consisting of *Pimpinella anisum*, *Laurus nobilis*, *Melaleuca leucadendron*, *Elettaria cardamomum*, *Pseudotsuga menziesii*, *Foeniculum vulgare*,
5 *Ferula galbaniflua*, *Pelargonium odoratissimum*, *Pelargonium radens*, *Pelargonium capitatum*, *Helichrysum augustifolium*, *Andropogon muricatus*, *Lavendula officinalis*, *Origanum majorana*, *Milissa officinalis*, *Citrus aurantium*, *Melaleuca viridiflora*, *Ravensara aromatica*, *Sassafras albidum*, *Tagetes erecta*, *Tagetes patula*, *Verbena officinalis* and *Thujopsis dolabrata*.
10
2. An acaricidal composition comprising monoterpene compounds selected from at least one group consisting of carvacrol, (+)-fenchol, geraniol, linanol, (-)-*cis*-myrtanol, *trans*-myrtanol, (-)-myrtenal, (-)-myrtenol, thujone, *cis*-verbenol, (-)-verbenone, menthone, eugenol,
15 menthol and indoline.
3. The acaricidal composition of claim 1 or 2, wherein the composition comprises said plant oils or monoterpene compounds in an amount of 0.01 to 40 % (w/w) in the amount of total composition.
20
4. The acaricidal composition of claim 3, wherein the composition comprises said plant oils or monoterpene compounds in an amount of 0.1 to 20 % (w/w) in the amount of total composition.
- 25 5. The acaricidal composition of claim 1 or 2, wherein the composition has acaricidal effect against mite.

6. The acaricidal composition of claim 5, wherein said mite is *Dermatophagoides pteronyssinus* or *Dermatophagoides farinae*.

7. The acaricidal composition of claim 1 or 2, wherein said composition is applied as a
5 form of volatile toxicants or fumigate.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/KR02/01183

A. CLASSIFICATION OF SUBJECT MATTER**IPC7 A01N 65/00**

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

A01N 65/00, A01N 57/14, C07F 9/165

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

NPS, PAJ, Deiphion

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	Jp 58-23606 A (SUMITOMO Chemical Co., Ltd) 12 February 1983 See the whole document	1-13
Y	Jp 58-55402 A (MITSUI TOATSU Chemical Co., Ltd) 1 April 1983 See the examples and claims	1-13
A	Jp 58-62104 A (MITSUI TOATSU Chemical Co., Ltd) 13 April 1983 See the examples and claims	1-13
A	Jp 59-65005 A (CHUGAI PHARMACEUT Co., Ltd) 13 April 1984 See the examples and claims	1-13
A	Jp 57-98203 A (RIKAGAKU KENKYUSHO) 18 June 1982 See the examples and claims	1-13
A	Jp 58-23606 A (SUMITOMO Chemical Co., Ltd) 12 February 1983 See the examples and claims	1-13

☐ Further documents are listed in the continuation of Box C.☒ See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

25 SEPTEMBER 2002 (25.09.2002)

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INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/KR02/01183

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
Jp 58-23606 A	12-02-1983	None	
Jp 58-55402 A	01-04-1983	None	
Jp 58-62104 A	13-04-1983	None	
Jp 59-65005 A	13-04-1984	None	
Jp 57-98203 A	18-06-1982	None	
Jp 58-23606 A	12-02-1983	None	